Neutrons attenuation on composite metal foams and hybrid open-cell Al foam

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Abstract

A comprehensive investigation of monochromatic neutron attenuation effectiveness for close-cell composite metal foams (CMFs) and open-cell Al foam infiltrated with variety of second phase materials is presented using both experimental and theoretical methods. The experimental results indicated higher neutron flux reduction in open-cell Al foam with fillers compared to the close-cell CMFs due to their large percentage of low Z elements such as hydrogen, boron and carbon, with superior neutron attenuation performance, in their filler materials. The main factor controlling the shielding effectiveness of steel-steel CMFs is found to be the ratio of the thickness of the sphere wall to the sphere radius while the intermetallic phases in the matrix of Al-steel CMFs seem to have a major role on their shielding properties. Successful models that link the observed material properties and microstructure have been developed using Monte Carlo N-Particle Transport Code (MCNP) to verify the accuracy of the experimental results. Close-cell CMFs were proposed through three different sphere arrangements: simple cubic, body center cubic and face center cubic, whereas open-cell Al foam with fillers was represented by creating a three-dimensional structure using periodic unit cell through two approaches. The simulation results were found to be in good agreement with the experimental values. This research indicates the potential of utilizing light-weight close-cell CMFs and open-cell Al foam with fillers as nuclear shields replacing conventional materials to achieve a specified shielding level with additional benefits of excellent energy absorption and thermal isolation.

Keywords

Metal foam; Neutron attenuation; Monte Carlo simulation; Areal density; Radiation shielding;

1. Introduction

Radiation shielding is commonly applied at nuclear facilities to attenuate the background of ionization radiations to a minimum level for creating a safer workplace, meeting regulatory requirements and maintaining high quality performance. The conventional radiation shielding methods have a number of drawbacks: heavy concrete contains a high amount of elements that are not desirable for an effective shielding such as oxygen, silicon, and calcium [1]; a well known limitation of lead is its low machinability and toxicity, which is causing a major environmental concern. Therefore, an effective and environmentally friendly shielding material with increased attenuation and low mass density is desirable.

Close-cell composite metal foams (CMFs) and open-cell Al foam with fillers are promising materials in this regard. Close-cell CMFs possess several suitable properties that are unattainable by conventional radiation shields such as low density and high strength for structural applications, high surface area to volume ratio for excellent thermal isolation with an extraordinary energy absorption capability [2, 3]. Open-cell foam is made up of a network of interconnected solid struts, which allows gas or fluid media to pass through it [4]. This unique structure provided a further motive to investigate its application as radiation shields by infiltrating original empty pores with high hydrogen or boron compounds, which are well known for their excellent neutron shielding capability. The resulting open-cell foam with fillers will not only exhibit light-weight and high specific surface area, but also possess excellent radiation shielding capability and good processability [5].

In our previous attempt, close-cell CMFs and open-cell Al foam with fillers were investigated against gamma rays, and it has demonstrated superior shielding effectiveness of close-cell CMFs at energy levels below 0.662 MeV [6]. Inspired by this understanding, we propose that high-Z elements in close-cell CMFs and low-Z elements in open-cell Al foam with fillers would endow these novel materials with excellent neutron shielding performance.

In the present work, close-cell CMFs were manufactured with either 2.0, 4.0 or 5.2mm steel hollow spheres embedded in Al or stainless steel matrix. These samples were tested against Neutron Powder Diffractometer beam along with another set of open-cell Al foam infiltrated with petroleum wax, borated polyethylene, water, or borated water. The goal of the study was to determine the feasibility of employing these high-performance materials in nuclear structures. In close-cell CMFs samples, the dependency of neutron transmission on sphere size and microstructure was emphasized, whereas in open-cell Al foam with fillers, the influences of sample thickness and filler materials on shielding effectiveness were explored. Monte Carlo N-Particle Transport Code version 5 (MCNP5) [7] was utilized to compare, verify, and validate the accuracies of the experimental results.

2. Materials Processing and Characterization

2.1. Materials and Sample Preparation

Aluminum A356 casting alloy (TriAlCo, Inc), stainless steel hollow spheres (Hollomet GmbH, Dresden, Germany), and 316L stainless steel powder (North American Hoganas High Alloys LLC) with particle size sieved to -325 mesh (95%) and -200/+325 mesh (5%) were used

in processing of close-cell CMFs. Three sizes of spheres with outer diameters of 2.0, 4.0, and 5.2mm were used for manufacturing close-cell CMFs. Steel-steel close-cell composite metal foams (steel-steel CMFs) comprising of steel hollow spheres closely packed in 316L stainless steel powder were manufactured through powder metallurgy technique. Fig 1 shows digital images of sectioned samples of the steel-steel CMFs produced with three sizes of spheres. Aluminum-steel composite metal foams (Al-steel CMFs) consisting steel hollow spheres and a solid aluminum A356 alloy matrix were processed through gravity casting technique. Fig 2 is cut section of Al-steel CMFs with three sphere sizes. Elemental compositions of hollow spheres are shown in Table 1 as well as compositions of both aluminum A356 alloy and 316L stainless steel matrix materials. More details of processing techniques are presented elsewhere [8, 9, 10, 11].

Open-cell Al foams (ERG Aerospace Corporation) was manufactured from Al 6101-T6 alloys with 5 pores per linear inch (PPI) of 97% porosity (ϵ_o) [12]. As indicated in Fig 3, open-cell Al foam was contained in an Al box (6061 Al alloy) and sealed after infiltrating the porosities with hydrogen-rich materials. As the result, each sample consists of three material layers. Top and bottom face sheets are made of 6061 Al alloy, and a hybrid middle layer is made from open-cell Al foam infiltrated with an additional filler: petroleum wax (IG Wax), borated polyethylene (5 wt% boron, Shieldwerx), water, or borated water (1 wt% boric acid). Chemical compositions of filler materials along with 6061 Al alloy and open-cell Al foam are presented in Table 2. More details of sample preparations can be found elsewhere [6].

Total eleven sets of samples were designed and tested to evaluate their shielding capabilities against Neutron Powder Diffractometer beam:

- Steel-steel CMFs (with either one of 2.0, 4.0, or 5.2mm spheres)
- Al-steel CMFs (with either one of 2.0, 4.0 or 5.2mm spheres)
- Aluminum alloy A356
- Open-cell Al foam + paraffin wax, sandwich composite (Open-cell Al foam + wax)
- Open-cell Al foam + borated polyethylene, sandwich composite (Open-cell Al foam + PE)
- Open-cell Al foam + water, sandwich composite (Open-cell Al foam + water)
- Open-cell Al foam + borated water, sandwich composite (Open-cell Al foam + borated water)

Aluminum A356 alloys are one of the most commonly used materials in nuclear industry, therefore, it was chosen as reference material in this study. All samples have been designed to have the same cross sectional area for testing (rectangular area 50.8mm x 89.3mm), and sample thickness is varied to provide areal densities of 2, 5 and 10g/cm². This allowed direct comparisons of attenuation capabilities of different materials at a constant shield weight. Physical properties of close-cell CMFs and open-cell Al foam with fillers are presented in Table 3.

2.2.Characterization

Samples for optical and scanning electron microscopy (SEM) analysis were prepared using a Buehler Isomet 4000 precision cutting saw. Thin slice of close-cell CMF samples were cut with a wafering blade rotating at a rate of 2500rpm and 1.0mm feeding rate. These samples

were surfaced using Buehler Automet 2 Power Head grinding and polishing stations. Grinding was conducted using a progression of 240, 600, 1200, and 4000 grit papers at a wheel speed of 90rpm for steel-steel CMFs and 70rpm for Al-steel CMFs. Each sample was then polished with 3µm diamond slurry, and followed by 1, 0.3, and 0.05µm alumina paste, using a wheel speed of 120rpm for steel-steel CMFs and 70rpm for Al-steel CMFs. Between each grinding and polishing stage, samples were carefully cleaned in an ultrasonic cleaner to prevent cross contamination.

Buehler Unimet Unitron 9279 microscope equipped a Hitachi KP-M1 CCD digital camera was utilized to obtain optical microscopy images in order to evaluate the microstructure of the sphere and matrix. SEM analysis was performed using Hitachi S-3200N environmental SEM equipped with energy dispersive X-ray spectroscopy (EDX) in order to investigate the more detailed microstructure of samples.

2.3.Density Analysis

Density of porous structure (ρ_p) can be calculated by:

$$\rho_p = \rho_s \varepsilon \quad (1)$$

where ρ_s is the density of the solid material and ε is the material porosity.

If the solid material contains N constituents, ρ_s can be obtained by rule of mixture in terms of weight fraction:

$$\frac{1}{\rho_s} = \sum_{i=1}^N \frac{w_i}{\rho_i} \quad (2)$$

where ρ and w are respectively the density, and weight fraction of constituent denoted by the subscript.

For close-cell CMFs, porosities (ε_c) of sphere wall and matrix were estimated by using open source software Image J version 1.47 [3, 13]. Optical images obtained from each sample was first turned into binary images with black and white color only, white dots represent the porosity inside selected region (either sphere wall or matrix). The software then estimated the porosity by measuring percentage of white dots in the given area. Ten measurements were undertaken for each sample and the average of the ten was used in the density calculation. Results were compared and verified with experimental measurements.

3. Neutron Transmission Measurements

A neutron interacts with the medium through two major processes: scattering and absorption [14]. The macroscopic cross section is the probability of a given reaction occurring per unit travel of the neutron, and is essential for providing basic data which determine transport

and attenuation of radiation in materials. The total macroscopic cross section of the interaction is equal to the sum of probabilities for each process by relation $\Sigma = \Sigma_s + \Sigma_a$ [14], Σ , Σ_s and Σ_a denote respectively the total, scattering and absorption macroscopic cross section of the medium.

The dead time of a detector is defined as the minimum time interval that two consecutive counts must be separated in order to be recorded as two different events. The relationships of the real beam intensity with the measured beam intensity can be expressed as [15]:

$$\Phi_t = \Phi_{mt} / (1 - \Phi_{mt} \tau) \quad (3)$$

$$\Phi_b = \Phi_{mb} / (1 - \Phi_{mb} \tau) \quad (4)$$

$$\Phi_o = \Phi_{mo} / (1 - \Phi_{mo} \tau) \quad (5)$$

where Φ_t , Φ_b and Φ_o denote, respectively, the transmitted, background and incident neutron intensity after dead time correction. Φ_{mt} , Φ_{mb} and Φ_{mo} represent measured beam intensity of the transmitted, background and incident neutrons, respectively. τ is the dead time (5 μ s). The neutron transmission can be obtained by the following equation [14]:

$$T = (\Phi_t - \Phi_b) / (\Phi_o - \Phi_b) = \exp(-\Sigma x) \quad (6)$$

where x (cm) is the sample thickness.

Neutron transmission measurements were carried out using the Neutron Powder Diffractometer beam available at North Carolina State University PULSTAR nuclear reactor [16]. At this facility, the thermal neutron intensities of approximately 2.7×10^{12} n/cm²/s is obtained at the entrance of beam tube #4 and approximately 0.64×10^5 n/cm²/s at the guide aperture with a reactor power of 1 MW. The beam is monochromatic and comprised of 0.035eV neutrons with a single energy correlating to a wavelength of 1.5 angstroms. The general experimental arrangement is shown in Fig 4. The whole detection system was held on an Aluminum 6061 table 83cm from the floor. The sample was placed at 36cm downstream from the guide aperture to allow for the greatest possible neutron flux. The neutron beam was limited to be 1.3cm in diameter by stacking of two Cd collimators with dimensions of 20x20x1cm³, another two stacking collimators with the same dimensions were placed 22cm downstream from the sample. These four collimators provide specific intensity and geometry to the neutron beam, meanwhile, eliminating the side scattered radiation from the beam line and enhancing its discrimination capability. A BF₃ detector (GE-Reuter Stokes model RS-P1-1506-202) was inserted into a B₄C shield tube which was used to decrease the counting of unwanted background radiation. The detector is 5cm in diameter by 46cm long. The filled gas is BF₃, with 90% ¹⁰B enrichment at a pressure of 20cm Hg. The detection system was placed 3cm downstream from the fourth Cd plate perpendicularly to the beam axis. The detector was mounted onto a preamplifier (ORTEC Model 142PC) and the output from the preamplifier was coupled to an amplifier (ORTEC Model 575) which provided signals to integral discriminator (ORTEC Model

421) to cut off low energy background peaks. The transmitted number of neutrons was displayed on the Timer and Counter (ORTEC Model 871). The detector and its electronics linearity was tested and checked before the course of experimental measurements by measuring the detector count rate at reactor power at every 100kW intervals from 1 to 10^3 kW. The data shows excellent linearity and stability. The transmitted (Φ_{mt}) neutron intensities were recorded in a 600s interval which was reasonable enough to obtain a statistical uncertainty in the range of 0.42% to 0.47%. Statistical uncertainty was determined by $1/\sqrt{N}$ (N is number of counts). The background radiation (Φ_{mb}) was subtracted from each measurement to correct for the contribution of nuclear radiation from some extraneous sources at PULSTAR reactor (i.e., Neutron Imaging source at beam tube #5, and Intense Positron source at beam tube #6), or radiation from the environment (cosmic rays, building materials, etc). The background measurement was conducted by placing a 5cm thick paraffin brick at the sample position, and recorded in a 600s interval. The neutron transmission was calculated using Equation (6) after acquiring the corresponding open beam (Φ_{mo}) at a constant time of 10s. Open beam was counted repeatedly between each sample measurement within a very stable error of $\pm 2\%$, therefore, all the measurements were performed without a beam-monitoring detector. Experimental results were compared with theoretical values calculated by MCNP5 developed at the Los Alamos National Laboratory [7].

4. Results and Discussions

4.1. Effect of sample thickness on neutron shielding

Fig 5a shows neutron transmission as a function of thickness in all eleven sets of samples. It can be clearly seen that the transmission decreases with increasing of sample thickness. Regardless of the type of samples in close-cell CMFs, approximately 20%, 40%, and 70% of the incident neutrons were attenuated at areal densities of 2, 5 and 10g/cm^2 , respectively. In Fig 5b, open-cell Al foam with fillers shielded up to 96% of the incident neutron flux at an areal density of 2g/cm^2 . With further increasing of the sample thickness, neutrons were 100% attenuated. The strong dependency of transmission on the sample thickness can be explained by the relation $T = \exp(-\Sigma x)$ (Equation 3) with transmission decreasing exponentially with increasing of the sample thickness.

4.2. Effect of type of samples on neutron shielding

At each given thickness, open-cell Al foam with fillers clearly dominates close-cell CMFs in terms of neutron flux reduction. This is attributable to the presence of higher contents of hydrogen and boron, which are known as effective neutron absorbers [4]. As summarized in Table 2, open-cell Al foam infiltrated with wax, PE, water, or borated water contain respectively 14.8%, 11.6%, 11.2%, and 11.1% (wt) of hydrogen. The presence of low Z element, hydrogen, serves as excellent neutron absorbers to capture thermal neutrons and endows open-cell Al foams with fillers with superior neutron attenuation performance.

Unlike open-cell Al foam with fillers that scattering is the predominant interaction mechanism, in close-cell CMFs the dominating mechanism is “radiative capture”. This is due to the fact that close-cell CMFs contain large amount of high-Z elements: Mn, Mo and Fe, which are very effective for such neutron absorption. As mentioned above, steel-steel CMF and Al-steel

CMF were produced by embedding the steel hollow spheres in stainless steel and Al matrix, respectively. Regardless of sphere size, as shown in Fig 5a, steel-steel CMFs perform better in terms of neutron attenuation than that of Al-steel CMFs. This is due to higher content of high-Z element Fe in steel-steel CMF matrix. Neutron absorption cross-section of Fe (2.59b) used in steel-steel CMFs matrix ranges over ten times than that of Al (0.213b) in Al-steel CMFs matrix. As a result, neutrons were absorbed more efficiently in steel-steel CMFs than that of Al-steel CMFs.

It is worth noting that both close-cell CMFs and open-cell Al foam with fillers perform better on neutron attenuation than that of Aluminum A356 with additional advantage of lightweight and thermal isolation, which are key technological requirements for the development of practical radiation shields.

4.2.1. Close-cell CMFs

4.2.1.1. Steel-steel CMFs

Effect of sphere size on neutron attenuation: The neutron transmission of steel-steel CMFs with different sphere sizes as a function of sample thickness is represented in Fig 6a. It was found that the transmission curves are closely overlapping with each other for all samples with different sphere sizes. This is due to the fact that the ratio of sphere wall thickness (t) to the radius of sphere (R) is maintained constant for all sphere sizes. As a result, the total amount of air inside the spheres is balanced out by total amount of sphere wall and matrix material in all samples despite of their different sphere sizes. As given in Table 4, all spheres with diameters of 2.0, 4.0 and 5.2mm possess a similar t/R ratio. At the same time, since the matrix and spheres are from similar material, the difference in the contact area of spheres and matrix in samples with different sphere sizes are not having any major effect in their shielding behavior. Therefore, attenuation efficiency of all steel-steel CMFs is relatively independent of sphere size.

Effect of microstructure on neutron attenuation: SEM observation was carried out to investigate the effect of microstructure on neutron attenuation. Figs 7a- 7c give SEM images of steel-steel CMF samples produced with 2.0, 4.0, and 5.2mm spheres. It can be seen that sphere walls and matrix are bonded well and no precipitation is presented. As indicated in Fig 7b, all spaces between 4.0mm spheres were completely filled with steel powder, the steel matrix is in close contact and well bonded to sphere walls. In Figs 7a and 7c, micro-porosities in the matrix can be observed as a result of lower steel powder compaction. Due to higher surface roughness of 5.2mm spheres [3], a small amount of debonding can be seen in Fig 7c between the spheres and the matrix in the samples made with 5.2mm spheres. Since the porosities in steel matrix are in the range of microscale, and the amount of debonding between spheres and matrix is relatively small. It has been studied and calculated in our previous study that the total voids in the Al matrix have an effect on density change of less than 1% [11]. Therefore, their contribution on neutron shielding performance seems to be negligible.

The porosities in matrix and within sphere wall for each sample were evaluated using imaging J [13]. The densities were then calculated by using equation (1), in which ρ_s of both sphere wall and the matrix was considered to be 8.0g/cm³. Details are listed in Table 5. These

1 results will be used later in modeling the shielding behavior of steel-steel CMFs against
2 neutrons.

3 4.2.1.2. Al-steel CMFs

4 **Effect of sphere size on neutron attenuation:** The transmission of Al-steel CMFs with
5 different sphere sizes as a function of thickness is represented in Fig 6b. It can be seen that
6 4.0mm Al-steel CMF clearly dominates 2mm and 5.2mm Al-steel CMFs in terms of neutron
7 attenuation while the transmission curves of 2.0mm and 5.2mm Al-steel CMFs are closely
8 overlapping. This may be attributable to three possible factors: spheres removal from sample
9 surfaces, voids between spheres and intermetallic phases within Al matrix. The first factor is
10 only a surface effect, while the last two factors are more of volume effect.

11 During the process of casting Al-steel CMFs, molten Al rose up to the top of the mold
12 and filled the interstitial space between the spheres. By decreasing the sphere size, unfilled
13 portion will increase due to the viscosity of the molten Al. Fig 2 shows the voids between
14 spheres owing to lack of aluminum penetration. The unfilled portion may have the effect of
15 decreasing the neutron attenuation, which suggested that the shielding efficiency would decrease
16 by decreasing the sphere size. However, this assumption is not inconsistent with the
17 experimental data indicated in Fig 6b. This result implies that the presence of voids in Al matrix
18 exhibits relatively small effect on neutron reduction due to the fact that the voids are in
19 microscale range and their contribution on neutron shielding performance seems to be negligible.

20 Spheres removal was mainly caused during sample preparation process, such as cutting
21 and ultrasonic cleaning. As indicated in Fig 2a and Fig 8, missing spheres are presented at the
22 surface of 2.0mm Al-steel CMFs as a result of a lack of fluidity in the molten Al. This may be
23 responsible for the reduction of neutron attenuation efficiency in 2mm Al-steel CMFs. The
24 5.2mm spheres had larger sphere shrinkage and surface roughness [3], which cause debonding of
25 spheres from matrix and their removal during samples preparation as well, shown in Fig 2c. As a
26 result, 5.2mm sphere CMFs lost spheres that caused reduction of neutron attenuation similar to
27 the 2mm sphere CMFs. Overall, 4mm Al-steel CMFs show an obvious advantage in shielding
28 neutrons as a result of the good interfacial bonding between spheres and Al matrix. Since spheres
29 removal is a surface effect, it was speculated that its influence on shielding efficiency would be
30 more severe in lower areal density samples than higher one. However, in comparison with 2mm
31 and 5.2mm Al-steel CMFs, 4mm Al-steel CMFs are measured to be 5%, 16%, and 32% more
32 efficient at areal densities of 2, 5 and 10g/cm², respectively. Therefore, further effort is needed to
33 fully understand the mechanism behind this phenomenon. As shown in Table 6, Al-steel CMFs
34 made of different sphere sizes: 2mm, 4mm, and 5.2mm, were calculated to contain the same
35 volume percentages of sphere wall material (15%), Al matrix material (41%) and air (44%).
36 Considering 4mm Al-steel CMFs possessed much higher intermetallic phases than that of 2mm
37 and 5.2mm CMFs, and the amount of such intermetallic phases increased with increasing the
38 areal density. It might be safe to infer that there would be a connection between shielding
39 effectiveness and intermetallic compounds, such as percentage, morphology or chemical
40 compositions. For this purpose, SEM analysis was performed in order to investigate the
41 microstructure of samples, and gain a comprehensive understanding of interaction mechanism.

Effect of microstructure on neutron attenuation: In Al-steel CMFs, using dissimilar materials in spheres and matrix resulted in the formation of intermetallic phases during solidification process [9]. It has been reported that intermetallic phases contain high-Z elements, such as Fe and Cr, diffused from the spheres into the aluminum [9], which may offer advantage in enhancing neutron absorption. Shown in Figs 9a-9c are SEM images of Al-steel CMFs produced with 2.0, 4.0, and 5.2mm spheres. Figs 9d-9f are respectively SEM images taken from marked area (dash line) of Fig 9a-9c at higher magnification with sphere wall-matrix intermetallic layer highlighted. It can be seen from 4.0mm Al-steel CMFs samples, Figs 9b and 9e, that three major kinds of precipitates were formed due to interdiffusion of elements between spheres and matrix: interface layer around sphere wall, plate shape and needle shape precipitation distributed within the matrix. The intermetallic layer has been observed to be more predominant in 4.0mm Al-steel CMFs ($110.89\mu\text{m} \pm 10.20\mu\text{m}$ thick) than in 5.2mm samples ($10.01\mu\text{m} \pm 0.20\mu\text{m}$ thick). In the SEM images of 5.2mm Al-steel CMFs sample (Figs 9c and 9f), the lower contact area between spheres and matrix and resulted less diffusion flux between the two greatly decreased the formation of intermetallic layers around sphere wall as well as plate shape and needle shape precipitations in the matrix. Owing to the lower percentage of intermetallic phases, 5.2mm Al-steel CMFs is less efficient in attenuating neutrons as compared to 4.0mm Al-steel CMFs. 2.0mm Al-steel samples were expected to exhibit a better neutron attenuation behavior due to their higher sphere-matrix interfacial areas. However, higher percentage of voids in 2.0mm Al-steel CMFs lowered the contact area between spheres and matrix causing the formation of a thinner intermetallic layer at their interface and lower percentage of plate shape and needle shape precipitation in the matrix (Fig 9a and 9d). All of the above may have resulted in the decrease in the shielding efficiency of 2.0mm Al-Steel CMFs. This result indicated that the dominant contribution on neutron shielding of Al-steel CMFs may be related to the formation of intermetallic phases around spheres and within the matrix. Further studies will be needed to fully understand the mechanism behind the phenomenon. The effect of intermetallic phases on radiation shielding will be reported in our future studies.

Chemical compositions of all intermetallic phases and their corresponding densities were calculated through equation (2) are given in Table 7. Image J was utilized to evaluate the dimensions of features in the microstructure as well as the fraction of each intermetallic phase presented in Al matrix, and details are summarized in Table 8. These results will be used later in modeling the shielding behavior of steel-steel CMFs against neutrons.

4.2.2. Open-cell Al foam with fillers

After introducing filler materials, such as petroleum wax, borated polyethylene, water, or borated water into foam porosities, the resulting open-cell Al foam possesses not only the excellent thermal isolation compared to bulk Al, but superior neutron shielding effectiveness from the hydrogen-rich filler materials. For a neutron of kinetic energy E encountering a nucleus of atomic weight A , the average energy loss can be expressed as $2EA/(A+1)^2$. The average energy loss reaches its largest value of $E/2$ by using hydrogen ($A=1$) [Error! Bookmark not defined.]. As shown in Fig 5b, open-cell Al foams with various fillers exhibited strong hydrogen dependency on attenuating neutrons. Open-cell Al foam + wax are measured to be more efficient than open-cell Al foam + water due to higher hydrogen content. It should be noticed that open-cell Al foam + PE with a hydrogen content of 11.6 wt% offers better neutron shielding

effectiveness than that of open-cell Al foam + wax with a hydrogen content of 14.8 wt% (Table 2). This may be attributed to the presence of an additional 5 wt% of boron in PE (Table 2). After being scattered by a nucleus, the neutron may be absorbed or captured by an absorber. Boron is one of the most commonly used neutron absorbers owing to its large absorption cross section [Error! Bookmark not defined.]. Similarly, open-cell Al foam + borated water presented an improvement on neutron shielding compared to open-cell Al foam + water as a result of the additional of only 0.1 wt% boron content.

5. MCNP Simulation

Simulation was performed in order to test the accuracy of the experimental results, and it was conducted through Monte Carlo N-Particle Transport Code version 5 (MCNP5), which is a general-purpose transport code for modeling the interaction of radiation with matter. In MCNP5 codes, the experimental setup follows narrow beam transmission geometry as described in Section 3. Tally F4 was used to obtain simulation data. This tally scores neutron flux in the detector cell. Simulations were performed with 10^6 histories. All the data obtained by MCNP5 were reported with less than 0.5% error.

5.1. Modeling of Close-cell CMFs

A 3D model was built by using MCNP5 code to evaluate neutron interaction with close-cell CMFs. The hollow sphere packing density of close-cell CMFs was reported to be 0.59 [2], which is classified as a random loose packing. In order to simplify the complex structure of close-cell CMFs, sphere packing arrangement was represented through three structures: simple cubic (SC), body centered cubic (BCC) and face centered cubic (FCC), with packing factor of 0.52, 0.68, and 0.74, respectively. Lattice parameter of each structure can be then calculated through $a_{SC} = 2R$, $a_{BCC} = 4R/\sqrt{3}$, and $a_{FCC} = 4R/\sqrt{2}$, R is sphere outer radius. While the steel-steel CMFs is easier to model, Al-steel CMFs is much more complicated due to the presence of different sphere wall and matrix materials, the intermetallic layer around the sphere wall as well as plate shape and needle shape precipitation which needed to be incorporated into the model. As a result, while R is only the outer radius of spheres in steel-steel CMFs, it is modified to be the sum of sphere outer radius plus the thickness of intermetallic layer in Al-steel CMFs. Plate shape and needle shape precipitations were represented by respectively cubical and cylindrical elements that are evenly distributed within Al matrix. The size and shape of these phases were estimated through image J and summarized in Table 9. The material properties for various components in the model can be found in Table 1 to Table 8.

Neutron transmission curves for close-cell CMFs with sphere arrangements of SC, BCC, and FCC predicted by Monte Carlo code for all samples are shown in Fig 10, and compared to corresponding curves based on the experimental results. It is interesting to find that shielding behaviors exhibit strong dependency on the model assumptions. In steel-steel CMFs, MCNP5 simulations predict that the SC structure is the most efficient sphere arrangement. This is attributed to the fact that SC structure has lowest packing density, which is a reflection of lowest percentage of air in the structure, and thus results in improving the shielding efficiency. It can be

clearly seen from Fig 10a-c that experimental values fall in between the SC and BCC theoretical curves which is a good agreement with the model prediction. A completely opposite behavior was observed in the case of Al-steel CMFs, as shown in Fig 11. The FCC structure possessed the highest attenuation efficiency. This can be attributed to its higher sphere packing density and resulted sphere-matrix interfacial area, which is causing the formation of greater amount of intermetallic precipitation along the sphere wall and within the matrix. As expected, the experimental curve lies in between SC and BCC curves, and are in accordance with our prediction. This study suggests that the presence of intermetallics can have an important effect on shielding behavior of materials.

5.2. Modeling of Open-cell Al Foam with Fillers

Open-cell Al foams investigated in this study is made of Al 6101-T6 alloys with 5 PPI of 97% porosity (ε_o) [12]. Fig 12 shows the digital image of open-cell Al foams. Due to the geometrical complexity and the random orientation of the solid phase of the porous medium, the real geometry of foams is not easy to characterize unless employing geometric idealization by using periodic unit cell. When the structure is periodic, the overall foam is represented by repeating that of a single unit cell, for which two schematic representations have been proposed in this study. In the first approach (Fig 13a): the periodic foam geometry is created by three equivalent cylindrical ligaments intersecting in three mutually perpendicular directions. In the second approach (Fig 13b): unit cell consists of four cylindrical ligaments in each of the body diagonal direction interconnected at the center of the cell. Lattice parameter (a_o) of the unit cell was measured to be 0.473cm, which match the calculated value for 5 PPI foam with an error of $\pm 6.8\%$ as well as the measured values that reported from literature [12] within an error of $\pm 7.5\%$. Diameter of cylindrical ligaments in Model-1 ($d_1=0.0534\text{cm}$) and Model-2 ($d_2=0.0351\text{cm}$) is thus a function of foam porosity, filler density and lattice parameter, which were calculated through equation (4) and (5), respectively:

$$d_1 = \sqrt{\frac{4(1 - \varepsilon_o)a_o^2}{3\pi}} \quad (4)$$

$$d_2 = \sqrt{\frac{(1 - \varepsilon_o)a_o^2}{\sqrt{3}\pi}} \quad (5)$$

Fig 14 shows the comparison of simulated neutron transmission curves with experimental data for (a) wax, (b) PE, (c) water and (d) borated water. It clearly shows a reasonable agreement between the experimental and the theoretical results. The relative errors between the experimental and model predictions are calculated to be in the range of 11.9% to 14.2% for Model-1 and 10.8% to 14.0% for Model-2. A discrepancy is typically found between analytical models and experimental measurements, which is attributed to the fact that open-cell Al foam is naturally non-periodic, non-uniform and anisotropic, which may lead to deviation of the model presented in this analysis from experimental data. Model-2 provides a slightly better result than that of Model-1, thus, more complexities are necessitated to obtain an improved prediction.

6. Conclusions

It appears that the shielding effectiveness of steel-steel CMFs is mainly controlled by the ratio of sphere wall thickness to the sphere radius, whereas, for Al-steel CMFs, there are three factors that must be taken into account in determining the radiation shielding efficiency: the most significant factor is the percentage of intermetallic phases, the second factor is spheres removal from sample surfaces, and the third factor is the incomplete filling of spaces between spheres with the molten Al. Such void formations will cause debonding of spheres from matrix and lowering the number of spheres at the surface of the sample. It is notable that while the latest is only a surface effect, the first two factors are more of volume effect. In open-cell Al foam with fillers, low-Z elements in the filler materials serve as effective monochromatic neutron moderators and absorbers that endow the structures with superior neutron attenuation effectiveness. Successful models that link the observed material properties and microstructure have been developed to predict the shielding efficiency of close-cell CMFs and open-cell Al foam with filler. This research provides useful insight on the correlation of neutron shielding effectiveness with structure and consequently supplies valuable information into our future study on developing multilayer radiation protective structures by combining close-cell CMFs and open-cell Al foam with fillers to achieve highest performance with minimum density and thickness.

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Figure 1: Digital images showing steel-steel CMFs produced with (a) 2.0mm, (b) 4.0mm and (c) 5.2mm spheres with 316L stainless steel matrix

Figure 2: Digital images showing Al-steel CMFs produced with (a) 2.0mm, (b) 4.0mm and (c) 5.2mm spheres with Aluminum A356 matrix. Arrows indicate the voids between spheres and missing spheres on sample surfaces.

Figure 3: Digital images showing open-cell Al foam + PE at areal density of 5g/cm^2 , with removable top face sheet and a middle layer infiltrated with PE

Figure 4: Experimental geometry for neutron spectrometry system with (a) neutron beam, (b) Cd collimators, (c) sample, (d) B_4C detector shielding, (e) BF_3 neutron detector, (f) Al supporting table, (g) neutron beam shielding

Figure 5: Neutron transmission as a function of sample thickness for (a) all samples, and (b) open-cell Al foam with fillers

Figure 6: Neutron transmission as a function of sample thickness for (a) steel-steel CMFs, and (b) Al-steel CMFs

Figure 7: SEM images of (a) 2.0mm, (b) 4.0mm and (c) 5.2mm steel-steel CMFs showing porosity difference in matrix and sphere wall, dashed line showing the wall thickness

Figure 8: SEM images of 2.0mm Al-steel CMFs showing missing spheres and voids in Al matrix

Figure 9: SEM images of (a) 2.0mm, (b) 4.0mm, and (c) 5.2mm Al-steel CMFs showing plate shape and needle shape precipitations, dashed line showing intermetallic layer. (d), (e), and (f) are respectively enlarged images from boxed area of (a), (b), and (c)

Figure 10: Comparison of experimental and theoretical transmission curves for (a) 2.0mm, (b) 4.0mm, and (c) 5.2mm steel-steel CMFs

Figure 11: Comparison of experimental and theoretical transmission curves for (a) 2.0mm, (b) 4.0mm, and (c) 5.2mm Al-steel CMFs

Figure 12: Digital image of open-cell Al foam supplied by ERG Aerospace Corporation with 5PPI

Figure 13: Representations of open-cell Al foam (a) Model-1 (b) Model-2

Figure 14: Comparison of experimental and theoretical transmission curves for open-cell Al foam with (a) wax, (b) PE, (c) water, and (d) borated water

Figure-1

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Figure-13

Figure-14

Table 1(a): Chemical compositions of different spheres in close-cell CMFs (wt%)

	2.0mm diameter	4.0mm diameter	5.2mm diameter
C	0.68	0.58	0.87
Mn	0.13	0.15	0.07
Si	0.82	1.14	0.34
Cr	16.11	17.34	17.09
Ni	11.53	12.28	12.60
Mo	2.34	2.28	2.12
P	--	0.009	--
S	--	<0.003	--
Cu	--	0.04	--
Co	--	0.02	--
Fe	balance	balance	balance

Table 1(b): Chemical compositions of matrix in close-cell CMFs (wt%)

	316L Stainless Steel Matrix	Aluminum A356.2 Matrix
C	0.030	--
Mn	2.00	0.28
Si	1.00	7.01
Cr	16.00-18.00	0.02
Ni	10.00-14.00	--
Mo	2.00-3.00	--
P	--	--
S	--	--
Cu	--	0.11
Co	--	--
Fe	balance	0.50
Mg	--	0.39
Ti	--	0.09
Zn	--	0.06
Al	--	balance

Table 2(a): Chemical compositions of filler materials (wt %)

	Petroleum wax	Borated polyethylene	Water	Borated water
H	0.148	0.116	0.112	0.111
B	--	0.050	--	0.002
C	0.852	0.612	--	--
O	--	0.222	0.888	0.887

Table 2(b): Chemical compositions of 6061 Al alloy and open-cell Al foam (wt %)

	6061 Al alloy	Open-cell Al foam
B	--	≤ 0.060
Cr	0.04-0.35	≤ 0.030
Cu	0.15-0.40	≤ 0.10
Fe	≤ 0.70	≤ 0.50
Mg	0.80-1.20	0.35-0.80
Mn	≤ 0.15	≤ 0.030
Si	0.40-0.80	0.30-0.70
Ti	≤ 0.15	--
Zn	≤ 0.25	≤ 0.10
other	≤ 0.15	≤ 0.10
Al	95.8-98.6	≥ 97.6

Table-3

Table 3(a): Physical properties of close-cell CMFs and Aluminum A356

	Designed areal density (g/cm ²)	Thickness (cm)	Volumetric density (g/cm ³)	Measured areal density (g/cm ²)	Difference of designed and measured areal density (%)
(2.0mm) Steel-steel CMF	2	0.77	2.50	1.93	-3.61
	5	1.91	2.50	4.78	-4.40
	10	4.03	2.36	9.50	-5.05
(2.0mm) Al-steel CMF	2	1.08	1.90	2.05	2.59
	5	2.62	1.97	5.17	3.48
	10	5.19	1.91	9.90	-0.98
(4.0mm) Steel-steel CMF	2	0.72	2.70	1.94	-2.79
	5	1.75	2.83	4.97	-0.58
	10	3.69	2.73	10.06	0.58
(4.0mm) Al-steel CMF	2	1.00	1.91	1.92	-4.05
	5	2.43	2.00	4.88	-2.39
	10	4.92	1.92	9.42	-5.77
(5.2mm) Steel-steel CMF	2	0.79	2.63	2.09	4.50
	5	1.81	2.71	4.91	-1.8
	10	3.65	2.71	9.91	-0.90
(5.2mm) Al-steel CMF	2	1.11	1.79	1.99	-0.50
	5	2.60	1.89	4.91	-1.80
	10	5.33	1.99	10.61	6.10
Aluminum A356	2	0.74	2.72	2.01	0.28
	5	1.86	2.70	5.03	0.66
	10	3.72	2.70	10.04	0.41

Table 3(b): Physical properties of open-cell Al foam with fillers

	Designed areal density (g/cm ²)	Thickness (cm)	Volumetric density (g/cm ³)	Measured areal density (g/cm ²)	Difference of designed and measured areal density (%)
Open-cell Al foam + Wax	2	1.54	1.31	1.32	1.96
	5	4.74	1.15	1.05	-0.98
	10	10.06	1.03	0.98	-1.23
Open-cell Al foam + PE	2	1.49	1.32	1.31	-2.49
	5	4.57	1.05	1.15	5.08
	10	9.69	0.98	1.03	-0.27
Open-cell Al foam + Water	2	1.37	1.35	1.84	-7.80
	5	4.14	1.20	4.98	-0.35
	10	8.77	1.15	10.05	0.53
Open-cell Al foam + Borated Water	2	1.37	1.35	1.84	-7.80
	5	4.14	1.20	4.98	-0.35
	10	8.77	1.15	10.05	0.53

Table 4: Comparison of variety of spheres used in close-cell CMFs

Sphere size (mm)	Sphere wall thickness t (mm)	Sphere outer radius R (mm)	t/R
2.0	0.104 ^a	1.02	0.1023
4.0	0.196 ^a	1.76	0.1111
5.2	0.244 ^a	2.59	0.0943

^a Ref. [3] The hollow spheres used in this study are from the same batches to the hollow spheres used in the reported literature [3], therefore, the sphere parameters are taken as the same.

Table 5: Physical parameters of steel-steel CMFs				
Steel-steel CMFs	Sphere wall porosity (%)	Sphere wall density (g/cm ³)	Matrix porosity (%)	Matrix density (g/cm ³)
2.0mm	8 ^a	7.36	32.61	5.39
4.0mm	6 ^a	7.52	0.33	7.97
5.2mm	4 ^a	7.68	42.24	4.62

^a Ref. [3]

Table-6

Table 6: Volume percentages of sphere wall material, matrix material and air in Al-steel CMFs

Samples	Sphere Wall Material (vol %)	Al Matrix Material (vol %)	Air ((vol %)
(2mm) Al-steel CMF	15.0	41.0	44.0
(4mm) Al-steel CMF	14.6	41.0	44.4
(5.2mm) Al-steel CMF	14.6	41.0	44.4

Table 7: Chemical Compositions^b and calculated solid density of intermetallic phases in Al-steel CMFs (wt %)

	Al	Si	Fe	Cr	Mn	Solid Density ρ_s (g/cm ³)
Intermetallic layer	58.52%	6.74%	27.57%	6.39%	0.78%	4.42
Plate shape	60.59%	8.17%	22.78%	7.51%	0.95%	4.23
Needle shape	72.60%	5.02%	21.53%	0.85%	0.00%	3.83

^b Ref. [7]

Table-8

Table 8: Physical parameters of Al-steel CMFs

Al-steel CMFs	Matrix porosity (%)	Matrix density (g/cm ³)	Thickness of intermetallic layer (μm)	Density of intermetallic layer (g/cm ³)	vol % of plate shape precipitation	Density of plate shape precipitation (g/cm ³)	vol % of needle shape precipitation	Density of needle shape precipitation (g/cm ³)
2.0mm	34.01	1.78	37.90±0.91	2.92	11.21	2.79	4.36	2.53
4.0mm	6.74	2.52	110.89±10.20	4.12	23.57	3.94	10.02	3.57
5.2mm	18.44	2.20	10.01±0.20	3.61	13.28	3.45	3.71	3.13

Table 9: Characteristics of SC, BCC, and FCC structures in MCNP5 modeling
(a) Steel-steel CMFs

		Lattice parameter a (cm)			
		SC	BCC	FCC	
Steel- steel CMFs	2.0mm	0.20	0.23	0.28	
	4.0mm	0.40	0.46	0.57	
	5.2mm	0.52	0.60	0.74	

(b) Al-steel CMFs

		Lattice parameter a (cm)			Plate shape precipitation (μm)	Needle shape precipitation (μm)	
		SC	BCC	FCC	Lattice parameter	Diameter	Length
Al-steel CMFs	2.0mm	0.21	0.24	0.30	42x42x42	6	96
	4.0mm	0.38	0.43	0.53	167x167x167	46	248
	5.2mm	0.52	0.60	0.73	14x14x14	4	57